Los Alamos SFA: Pu/Actinides in the Environment

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Earth and Environmental Sciences

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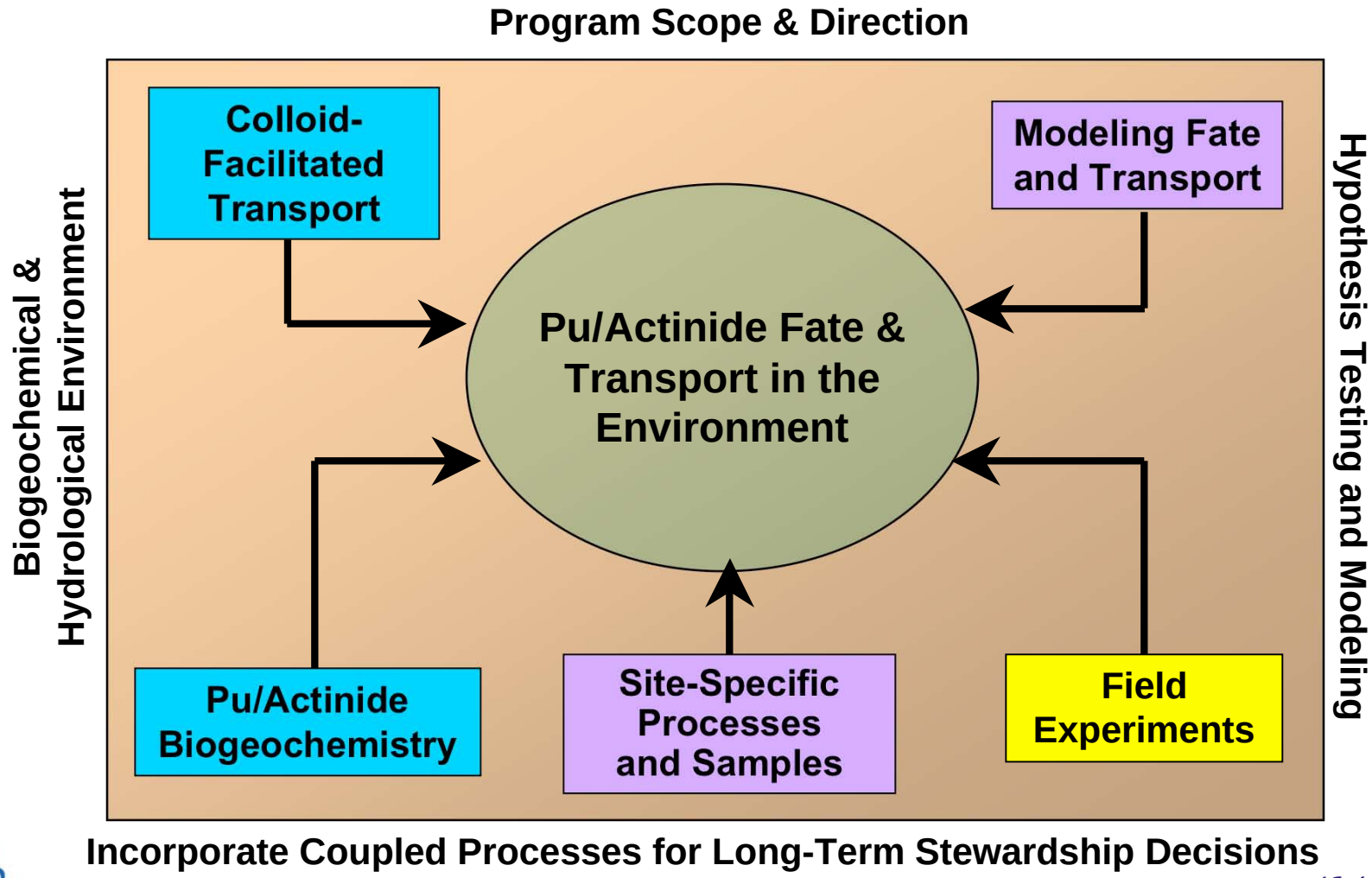
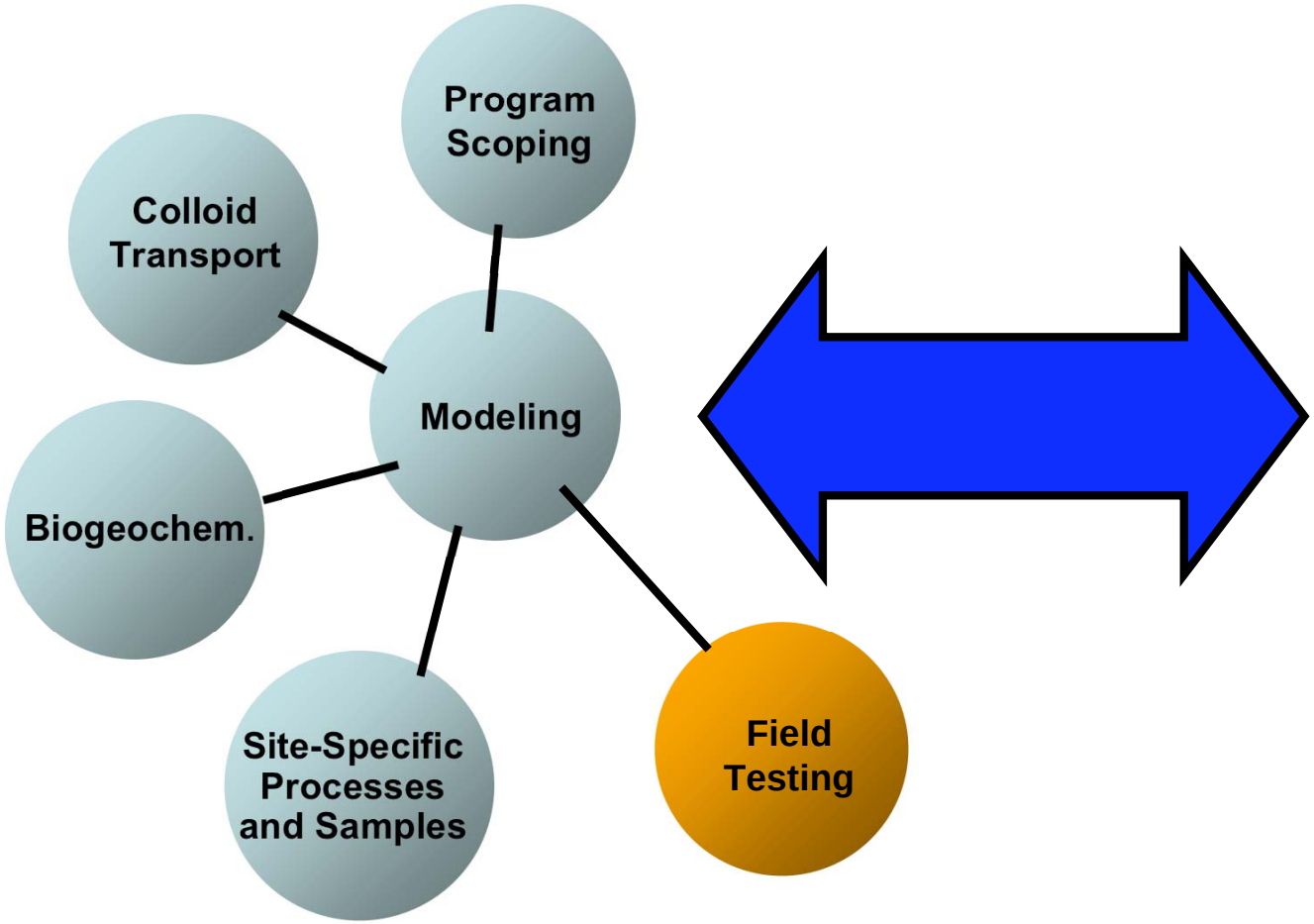
Pu/Actinide SFA Motives

- Provide Better Understanding of Pu/Actinides in the Environment, Develop Coupled Process Models for Long-Term Stewardship.
- Pu Inventories at Several DOE Sites
  - Los Alamos
  - Hanford
  - Idaho
  - ORNL
  - Savannah River
  - Rocky Flats
  - Nevada Test Site
- What Controls Pu/Actinide Fate in the Subsurface?
  - Intrinsic colloid formation, Pu association with natural colloids
  - Biogeochemical and hydrological processes that affect Pu subsurface fate and transport (e.g., Pu-organic forms; Pu redox; colloid formation)
- How Do We Predict Pu/Actinide Subsurface Transport?

Integrated LANL Research Program



Fundamental Understanding Leads to Application at Multiple Scales



Applications at Multiple Scales

- Field
- Column
- Pore
- Molecular

Fundamental Process Understanding

SFA Drivers: Public and Programmatic

- Significant Environmental Inventory
  - LANL, INL, Hanford, NTS, and maybe 11 other sites in US
  - Larger international inventory?
  - TRU and Nuclear Repositories (YMP, WIPP)
- High Public Visibility
  - Pu contamination, perceived or real, is scrutinized, publicized, and criticized by government agencies and public interest groups (300 hits for Pu on Concerned Citizens website alone).
- Complex Behavior, not Understood Well
  - Colloid-facilitated transport: a significant factor.
  - Biological processes affect Pu redox & speciation.
  - In the Lab: multiple redox states and distinct species.
  - In environment: sorption, move as or with colloids, or soluble species.

Technical and Scientific Research Questions

- What Controls Pu/Actinide Fate and Transport?
  - Source terms and source forms.
  - Range of redox and chemical conditions in actual subsurface environments.
  - Relevant biogeochemical & hydrological mechanisms in subsurface environments?
  - Colloid and colloid-facilitated transport; soluble Pu-complexes?
  - Data needs for models?
- How are Fate and Transport Predicted in Subsurface Environments?
  - Subsurface characterization needs?
  - Modeling approaches and appropriate scales?
  - Data needs for models?
  - Calibration of model predictions/simulations?

Guiding Ideas and Hypotheses

- We can bound the range of biogeochemical hydrological conditions from site characterization data.
- Colloids and complexed Pu are key forms for transport.
- Initial waste form and subsurface biogeochemistry determine Pu species formation and stability.
- Site-specific conditions and transients are key to understanding transport.
- Redox cycling is an important process in Pu fate and transport.

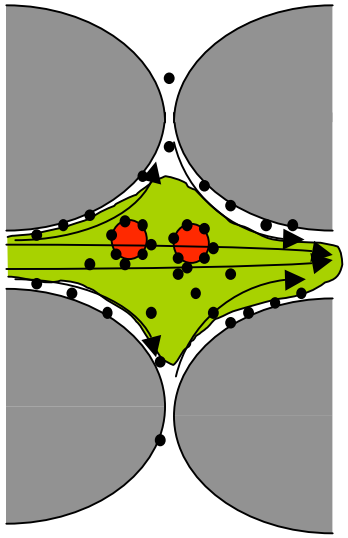
Coupling of colloid behavior, biogeochemical and hydrological processes will be integrated via modeling.

Bound Biogeochemical & Hydrological Environments using Site Characterization

- Riley & Zachara review (1992) and new site characterization since (e.g., EM Programs at LANL; work at Rocky Flats; RIBRA at INL)
- Applications of new technology to existing subsurface data; Information from IFCs
- Source form could be Pu oxides, aqueous Pu, associated/complexed Pu or ?
- Source terms could vary from less than pCi/kg levels to > 10^6 pCi/kg depending on site, processes, medium.
- Interaction between waste form and site-specific hydrology and biogeochemistry will be key in fate and transport.

Pu Transports as Colloids

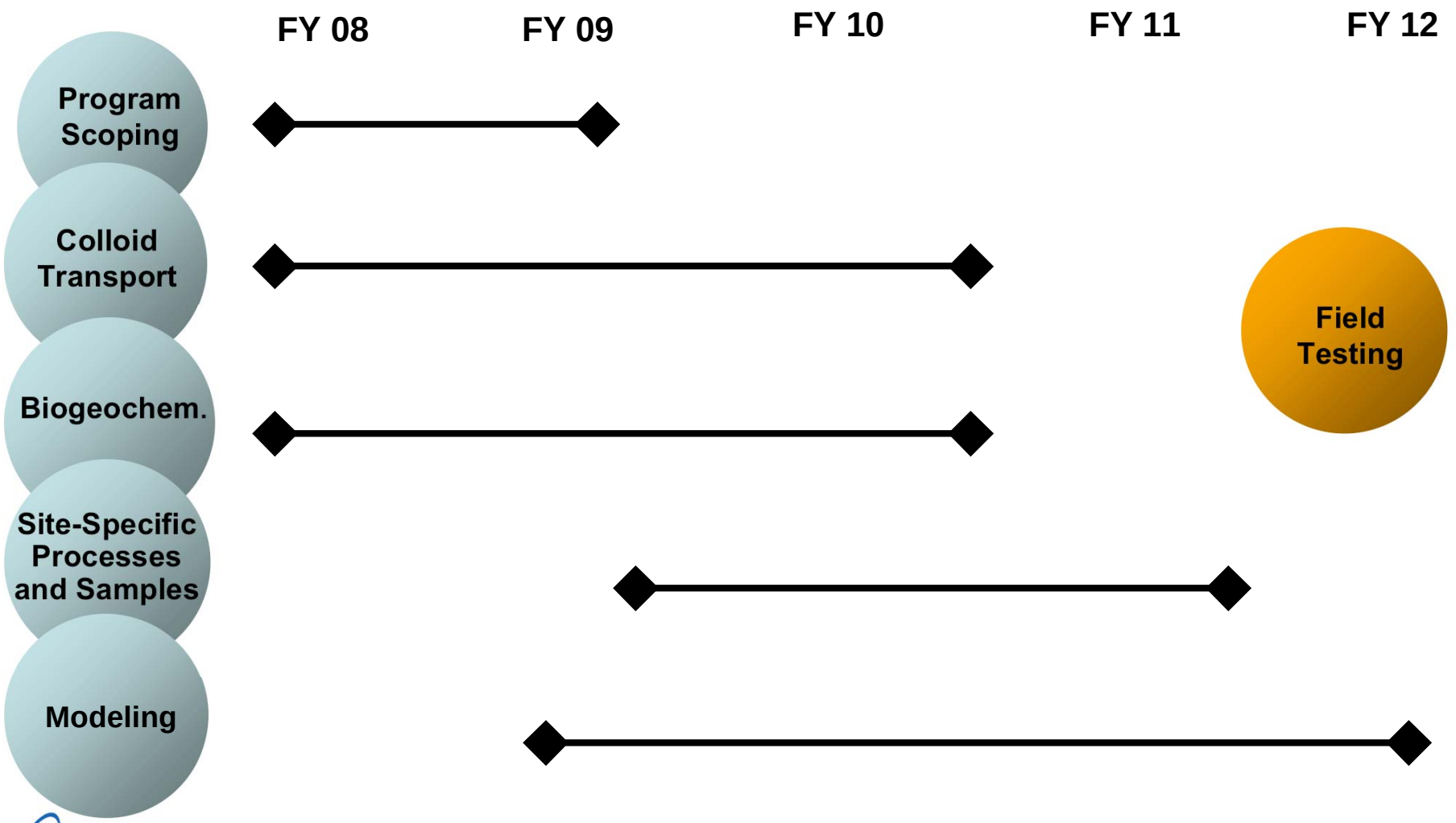
- Significant fraction of source term that transports is in colloidal or Pu-colloidal form.
- Colloids/Pu-colloids behave as “fast-lane vehicles” for transport in subsurface environments.
- Pore-scale colloid attachment and detachment kinetics are important.
- Pu partitioning between solution, immobile matrix, and mobile colloids.
- Sufficient quantity, limited colloid filtration, colloids stable



Pu Transport as Soluble Complexes

- Biogeochemical and hydrological conditions (e.g., redox, pH) favorable for formation of soluble Pu complexes (e.g., Pu-siderophores; Pu-EDTA; Pu-carbonates)
  - Conditions that stabilize oxidized Pu(V) and Pu(VI) (both more soluble than reduced Pu(III) and Pu(IV))
  - Under reducing conditions redox cycling between Pu(III) and Pu(IV) that leads to increased solubility.
- Pu partitioning between solution, immobile matrix, and mobile colloids.

LANL SFA Proposed Timeline



How Will We Accomplish This?

FY 08 → FY 12

Fundamental Understanding	Field-Derived Samples	Field Testing
Lab experiments	Lab results & methods as guides	LANL as Collaborator with IFC(s)
Experiments conducted with site-specific materials	Move into larger scales (column, larger)	Models of processes at different scales
Model experimental results	Bound experiments with biogeochemical-hydrological conditions.	
Molecular to column scale	Continue collaborations	
Collaborations (SSRL, INL)		

Review and Redirect

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Coupled Processes used for Long-Term Stewardship

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